Artificial Neural Networks Study on Prediction of Dielectric Permittivity of Basalt/PANI Composites

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Abstract-In the present study, the dielectric permittivity change of basalt (two type basalt; CM-1, KYZ-13) reinforced PANI composites were studied to determine the effects of PANI additivities (10.0, 25.0, 50.0 wt.%) at several frequencies from 100 Hz to 17.5 MHz by a dielectric spectroscopy method at the room temperature and artificial neural networks (ANNs) simulation. Also, the dielectric permittivity at 30.0 wt.% of PANI additivity was obtained by ANNs without experimental process. That process, a significant predictive instrument was produced which allows optimization of dielectric properties for numerous composites without substantial experimentation. It has been observed that PANI additivities decreased to dielectric constant of composites at low frequencies. Furthermore, the ANNs method have satisfactory accuracy for prediction of dielectric parameters.

Keywords: Artificial neural networks, dielectric permittivity, basalt, PANI, composite.

1. Introduction

There is growing interest in reinforcing polymer matrix composites with basalt reinforced polymers because of their moderate cost, high stiffness and strength, excellent corrosion and oxidation resistance, and heat resistance and thermal stability. Basalt is a very common volcanic rock, dark colored and comparatively rich in iron and magnesium, which is located at almost every country in the world. Basalt is used for a wide variety of purposes. It has been used in the rock industry to create industrial construction, highway engineering and building tiles for other purposes [1]. Polyaniline (PANI) is one of the most promising conductive polymers for technological applications due to its easy synthesis, high environmental stability, huge electrical conductivity, as well as a comparatively low cost [2, 3]. PANI is synthesized for specific applications like organic electronics [3], circuit component (such as varistors [4], super capacitors [3, 5]), electrochemical catalysis [5, 6], corrosion protection [7, 8] and sensors [9].

Artificial neural networks (ANNs) are becoming well-known because of their achievement where complicated nonlinear relationships occur amongst data. ANNs are biologically influenced computer programs created to simulate the way in which the human brain processes knowledge. ANNs collect this information through detecting the relationships and patterns in learned via experience and data. The broad use of ANNs is a result of their ability and versatility to model nonlinear systems without earlier information of an empirical model. They do not require a specific formulation of the physical or mathematical relationships of the undertaking issue. These give ANNs an advantage over traditional fitting methods for numerous application. [10].

In this work, the dielectric permittivity values of basalt reinforced PANI were determined at several frequencies from 100 Hz to 17.5 MHz by using experimental Impedance spectroscopy technique, and this data was used to develop an artificial neural networks model for next prediction of dielectric permittivity. The data samples were produced three different composite (10.0, 25.0, 50.0 wt.%). Then, we calculated the dielectric permittivity for 30.0 wt.% PANI additivity as a sample application of developed ANNs model.

2. Materials and Method

2.1. Materials

Basalt samples (two type basalt samples and coded CM-1, KYZ-13) obtained from different regions of Van in Turkey. Chemical analyses of the basalt samples are taken by X-ray fluorescence (XRF) instrument. Operating conditions of the XRF device of Philips PW-2400 were fixed at 50 mA and 60 kV. The results of chemical analysis of basalt samples are given in Table 1.

Table 1. Chemical composition of basalt samples [1].

Sample / Comp.	CM-1	KYZ-13
SiO_2	41.668	47.790
TiO_2	2.0800	1.3950
Al_2O_3	13.106	16.918
Fe_2O_3	13.823	10.878
MnO	0.1920	0.1630
MgO	9.7540	7.6190
CaO	10.602	11.357
Na_2O	5.2610	3.1370
K_2O	1.7370	0.5190
P_2O_5	1.7770	0.2240

Polyaniline (emeraldine base, average Mw ~5000) was purchased from Sigma-Aldrich. To investigate effects of PANI additive percentages (10.0, 25.0, 50.0 wt.%) on the dielectric properties of basalt mechanically modified by PANI. These composites were prepared by mixing of basalt samples with PANI and were molded by compression in a cold press at room temperature. Each one of these mixtures prepared as a pellet having 13±0.02 mm diameter, 0.500±0.050 g weight and 1.9-2.2 mm thickness.

2.2. Dielectric Measurements

The dielectric measurements were carried out with a two-point probe arrangement. Dielectric measurements have been performed by using an HP 4194A Impedance Analyzer in the frequency range from 100 Hz–15 MHz at room temperature with a high accuracy (0.17% typ.) In this work, the overall errors the dielectric measurements are 2.5% and the RMS amplitude of the instruments is ~500mV.

2.3. Artificial Neural Networks (ANNs)

ANNs are a mathematical model that can be defined as an interconnected set of nodes their connections. Also neural network is a network of neurons. The neurons are the pre-defined computational units that are grouped in sets of layers. Basically, there are three types of layers, input, output and hidden layer as seen in Fig. 1. The input layer receives data from the outside and transfers them to sub layers. The data comes from input layer is processed in the hidden layer by performing interconnection of neurons and sending the results to the neuron in the output layer.

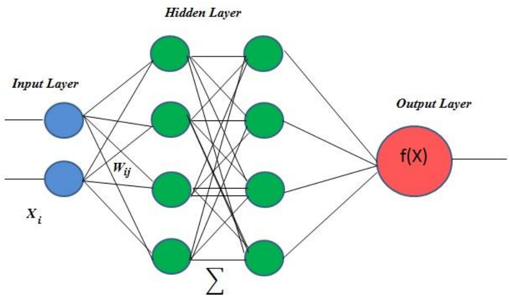

Fig. 1. The architecture of the ANNs algorithm.

The output layer submits the output data. The neurons perform weighted summation of the inputs that are applied to the networks calculated as [11];

$$Net_i = \sum_{j=1}^{N} X_{ij}.W_{ij} + n_i \tag{1}$$

Where W_{ij} is the weight between the i th neuron in the previous layer to the j th neuron. X_{ij} is the output of i th neuron and n_i is the bias value of previous layer.

The output of the i th neuron is calculated using a sigmoidal activation function as follows.

$$out_i = f\left(Net_i\right) = \frac{1}{1+e^{(-aNet_i)}} \tag{2}$$

Here, a is a constant used for controlling the slope of the semi-linear region [11].

There are two stages to process the network for an ANNs model. First of them is training and the other is testing. In the training stage, the network is training to predict an output based on the input. After that the training stage, the predicted output is compared with given output. This testing decides whether to stop or continue training of the network.

For the training of the network, a back propagation algorithm (BP) has been used. The Levenberg - Marquard (LM) algorithm [12, 13] is used to update the weights (W_{ij}) for minimizing the error function that defined as absolute difference between predicted output value and given actual value. In the LM algorithm, the Hessian matrix can be approximated as;

$$H = J^T J \tag{3}$$

and this approximation is used for the iteration of the weights;

$$x_{k+1} = x_k - \left[J^T J - \mu I\right]^{-1} J^T e \tag{4}$$

Where J is the Jacobian matrix, e is a vector of network errors, μ is the scalar learning rate and I is the identity matrix. When μ is large, this becomes gradient descent method. This method used as adaption function in this study.

The prediction accuracy performance of the networks was calculated using coefficient of determination (R) and relative error (RE);

$$R = \left(\frac{\sum_{i=1}^{N}(y_i - \bar{y}_i)(x_i - \bar{x}_i)}{\sqrt{\sum_{i=1}^{N}(y_i - \bar{y}_i)^2 \sum_{i=1}^{N}(x - \bar{x}_i)^2}} \right) \tag{5}$$

$$RE(\%) = \frac{|x_i - y_i|}{x_i} \times 100 \tag{6}$$

where y_i and x_i are predicted and actual values respectively.

3. Results

Dielectric spectroscopy is most reliable and powerful experimental technique which has been effectively used for the characterization of electrical properties of the polar materials [14]. The DS technique is based on analyzing the alternative current (a.c.) response of a materials to a sinusoidal perturbation, and subsequent calculation of dielectric parameters as a function of frequency and temperature [15].

The dielectric analysis gives the permittivity and conductivity of material as a complex permittivity $\varepsilon^*(\omega)$ parameter. Dielectric permittivity is a principal parameter that determines the coupling and distribution of electromagnetic energy during microwave and radiofrequency processing [15, 16]. The frequency dependence complex dielectric permittivity $\varepsilon^*(\omega)$ is given by

$$\varepsilon^*(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega) \tag{7}$$

Here, ω, $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are the angular frequency, the real part of complex permittivity (or called dielectric constant) and the imaginary part of the complex dielectric permittivity (or called loss factor), respectively. In dielectric material $\varepsilon'(\omega)$ represents the alignment of dipoles, which is the energy storage component and $\varepsilon''(\omega)$ represents the ionic conduction component [15].

In this study, experimental results of $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ for samples have been shown in Fig. 2-5. The increase of the doping percentage of PANI decreases the dielectric constant for both composites in low-frequencies. Moreover, the $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ have large values in the low-frequency region and decreases rapidly in 10.0 wt.% PANI composites.

Furthermore, the ANNs model is developed to predict the dielectric parameters ($\varepsilon'(\omega)$ and $\varepsilon''(\omega)$) of these composites. The experimental measurements are used to obtain the 200 data samples of dielectric parameters ($\varepsilon'(\omega)$, $\varepsilon''(\omega)$) for various concentration (wt.%) and frequency (ω). Concentration and frequency of 200 data samples are used as input and, dielectric parameters of corresponding them are used as output (target). 80% percent of the data are used as training data and the other 20% are the testing data. The training and testing data are selected us randomly.

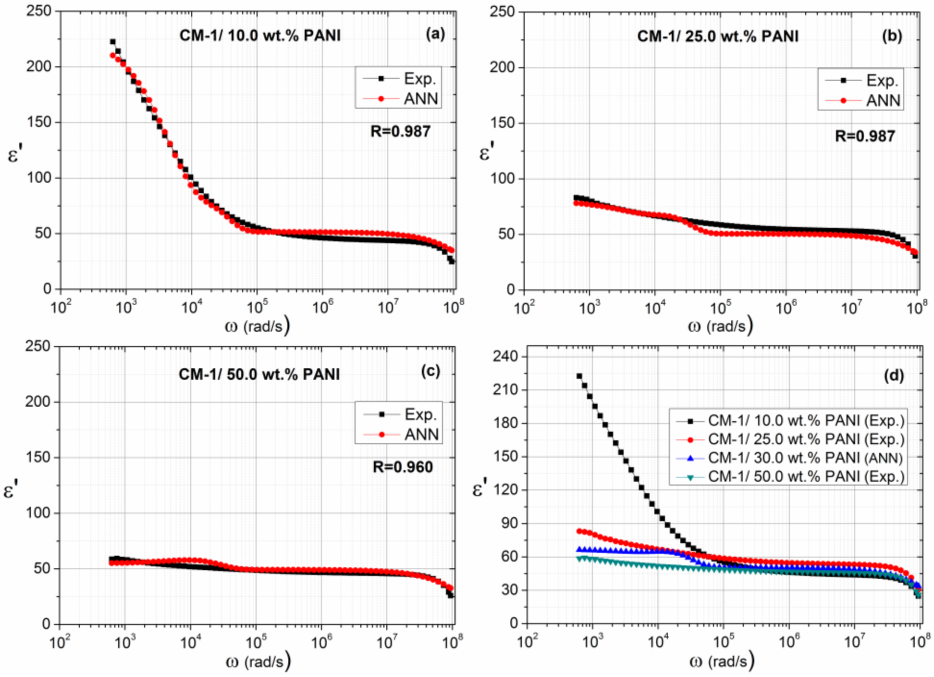

Fig. 2. Comparison of desired experimental results and predicted values of $\varepsilon'(\omega)$ for samples of **(a)** CM-1/ 10.0 wt.% PANI **(b)** CM-1/ 25.0 wt.% PANI **(c)** CM-1/ 50.0 wt.% PANI composites and **(d)** $\varepsilon'(\omega)$ prediction of CM-1/ 50.0 wt.% PANI composite

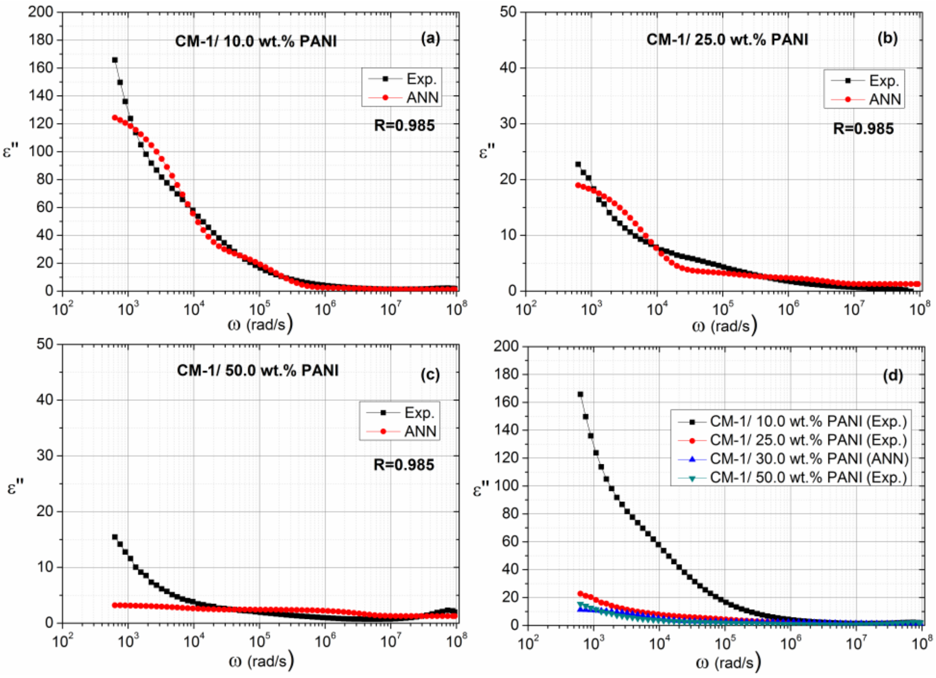

Fig. 3. Comparison of desired experimental results and predicted values of $\varepsilon''(\omega)$ for samples of **(a)** CM-1/ 10.0 wt.% PANI **(b)** CM-1/ 25.0 wt.% PANI **(c)** CM-1/ 50.0 wt.% PANI composites and **(d)** $\varepsilon''(\omega)$ prediction of CM-1/ 30.0 wt.% PANI composite

The comparisons of experimental results and output values predicted by ANNs of CM-1 are illustrated in Fig. 2-3. The figures show the variation of $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ with the frequency for various concentration (10.0 wt.%, 25.0 wt.% and 50.0 wt.%,). From these figures, it is seen that the high accuracy between experimental and simulated data. Therefore, it can be concluded easily that the outputs efficiently modeled by the ANNs model.

Also, it can be concluded from Fig. 4 and Fig. 5, that the KYZ-13 values can be efficiently modeled by the ANNs model similar to the CM-1.

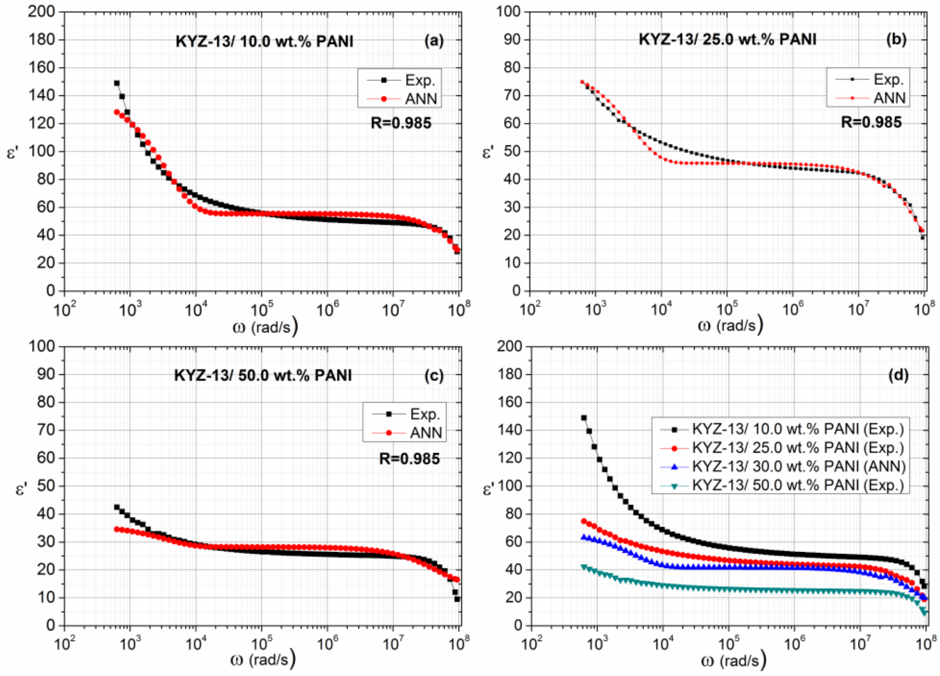

Fig. 4. Comparison of desired experimental results and predicted values of $\varepsilon'(\omega)$ for samples of **(a)** KYZ-13/ 10.0 wt.% PANI **(b)** KYZ-13/ 25.0 wt.% PANI **(c)** KYZ-13/ 50.0 wt.% PANI composites and **(d)** $\varepsilon'(\omega)$ prediction of KYZ-13/ 50.0 wt.% PANI composite

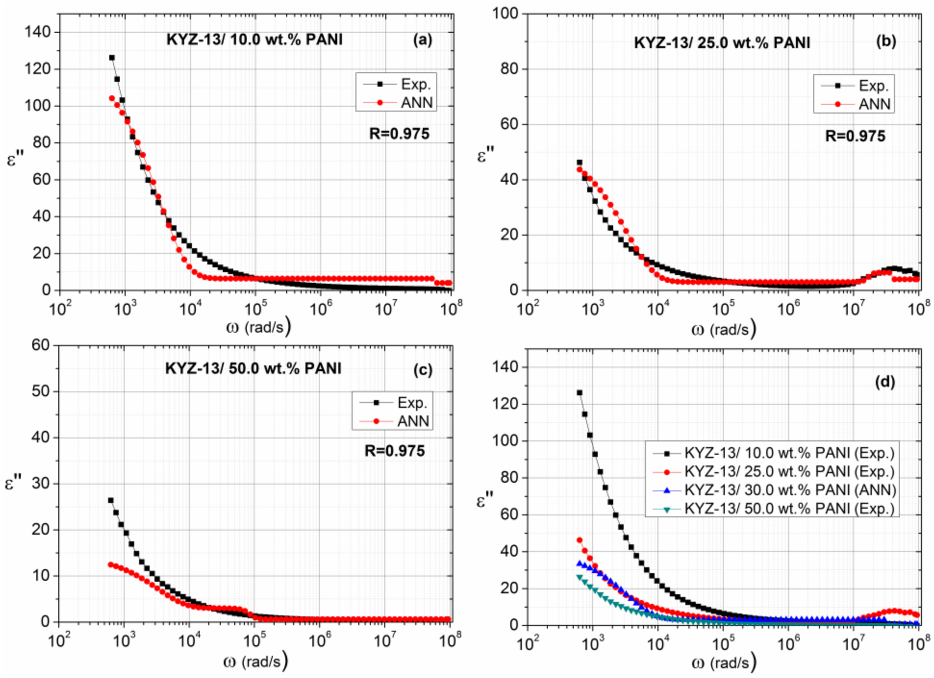

Fig. 5. Comparison of desired experimental results and predicted values of $\varepsilon''(\omega)$ for samples of **(a)** KYZ-13/ 10.0 wt.% PANI **(b)** KYZ-13/ 25.0 wt.% PANI **(c)** KYZ-13/ 50.0 wt.% PANI composites and **(d)** $\varepsilon''(\omega)$ prediction of KYZ-13/ 30.0 wt.% PANI composite

By using efficient training algorithm, the output values of $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ has been predicted for CM-1/ 30.0 wt. % PANI samples that were not measured in our experiments. The variation of predicted $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ values are figured in Fig. 2-d, Fig. 3-d, Fig. 4-d and Fig. 5-d. It can be seen that suitable output values can be obtained from ANN model for $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$.

The performance of the model on training and testing stages in terms of R is presented in Fig. 6 and Fig. 7.

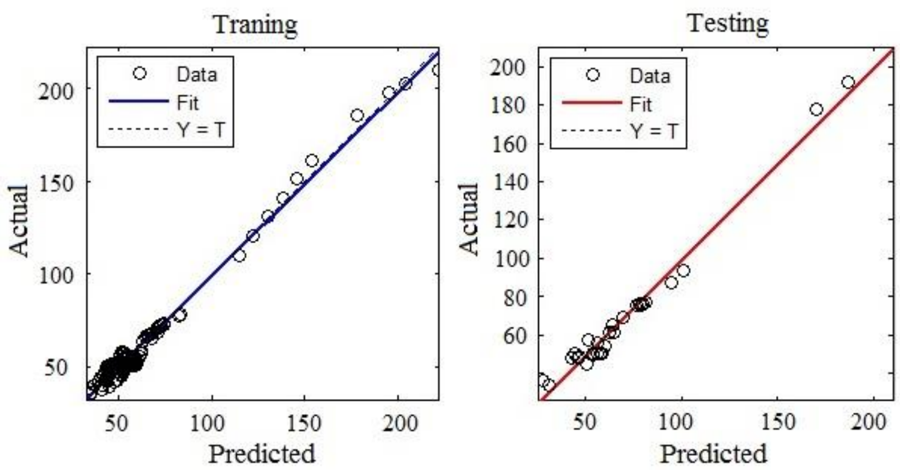

Fig. 6. Fit of ANNs model on **(a)** training data and **(b)** testing data of CM-1/PANI composites

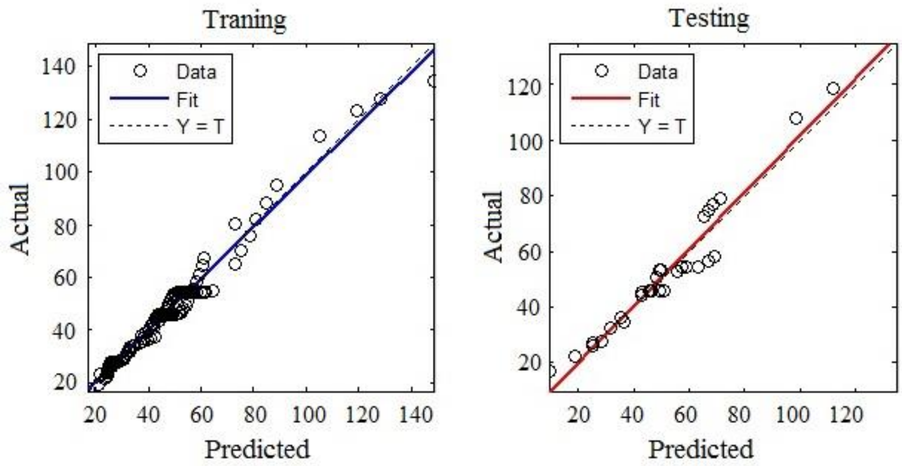

Fig. 7. Fit of ANNs model on **(a)** training data and **(b)** testing data of KYZ-13/PANI composites

The graphs show that the training algorithm worked well, and learned the non-linear relation between concentration, frequency and dielectric constant with high R value for both CM-1 and KYZ-13 samples. The R values of both training and testing are given in Table 2.

Table 2. Coefficient of determination (R) parameters of ANN for CM-1 and KYZ-13 composites

Sample / R	CM-1	KYZ-13
R (Training)	0.991	0.987
R (Testing)	0.989	0.972

4. Conclusion

This study represents the results of our investigation on the dielectric properties of two type Basalt/PANI composite using dielectric spectroscopy and an application of the ANNs method in the prediction of dielectric properties of basalt materials depend on additive percentage (%) and frequency (ω).

The results given in this paper prove that the ANN method has satisfactory accuracy for prediction of desired properties ($\varepsilon'(\omega)$ and $\varepsilon''(\omega)$). So the ANNs model can be used as an appropriate and practical method to compute dielectric properties of materials under various experimental conditions such as, concentration, frequency or temperature.

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